Russian Experience in Solid Radioactive Waste Processing: Achievements and Prospects

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Abstract—Plasma technology not only solves the problem of the elimination of the newly formed NPP operational waste, but also provides deep thermal processing of radioactive waste accumulated before or compacted in drums to make room for storage of solid radioactive waste.

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INTRODUCTION

The problem of disposal of low and intermediate level radioactive waste in the world is not finally solved. The main reason is the lack of technology, that proofed their feasibility and effectiveness, and therefore the absence of the facilities for the implementation of the proposed technologies for radioactive waste treatment.

One of the most common technologies to significantly reduce the amount of waste, followed by final burial of ashes, is the burning of radioactive waste in kilns. Only combustible components, separated from the waste are sent for incineration. The disadvantages of direct incineration of radioactive waste is the formation of hazardous during transportation, dusting and unsuitable for disposal of the product, the ash, concentrated radioactive isotopes [1]. Further conditioning of radioactive ash to convert it into a state safe for final disposal, requires additional industrial equipment and a number of technological operations (transportation ash for recycling, the introduction of additional material), which in some cases involves considerable energy expences.

Another disadvantage of the combustion related to the fact that the use of gas or liquid hydrocarbons with an excess of air for heating furnace and afterburning chamber results in the formation of large volumes of flue gases to be cleaned of harmful chemicals and radioactive substances before release into the atmosphere. For example, the combustion efficiency of the organic components of the waste on the grate is provided by two - threefold excess of supply air.

Plasma methods of direct processing of radioactive waste that can produce a product suitable for the transportation and disposal or long term storage have significant advantages. Compared with the traditional method of burning, plasma methods provide higher ratios of reducing waste volume and the volume of secondary waste generated, to obtain a product in the form of fused slag compound having a high mechanical strength and chemical resistance to aggressive environmental influences. Deterrent in the development of plasma processing technology of radioactive waste is a high degree of entrainment of volatile radionuclides (tens of percent), primarily Cesium-137 from melters and other high-temperature parts of plasma units.

GUP MosNPO "Radon" built and operates plasma facility "Pluton" which provides plasma processing of solid waste of complex morphology in the furnace shaft with performance of 200–250 kg/h producing a conditioned product in a single step with a high coefficient of waste reduction. Glass-like end product fused slag, is suitable for long-term storage or disposal at the landfill of conditioned radioactive waste. The

Table 1. Morphological composition of solid low- and intermediate active waste from Novovoronez NPP

	1
Component	Content, wt %
Paper, clothing, wood	55–65
Construction waste	15–35
Thermal insulation (glass- and rockwool)	10–20
Metal scrap	1–3
Ion exchange resins	3–5
Plasticate (PVC), rubber and polymers	2–3
Specific activity of ion exchange resins	$1.3 \times 10^7 \mathrm{Bq/kg}$
Specific activity of solid radioactive waste	$1.47 \times 10^6 \text{ Bq/kg}$

specific capital cost to build the plasma unit and operating costs for handling radioactive wastes by plasma does not exceed the cost of combustion, thus the unit is able to process simultaneously the waste fed for burning, melting, pressing and supercompacting. Plasma treatment of solid waste excludes the stage of cementing of incineration product, ash.

Facility process solid radioactive waste of mixed type, similar in morphological structure to solid radioactive waste from NPP. Recyclable waste includes, along with combustible materials (paper, wood, textiles, polymers) up to 40–50% of noncombustible components (construction debris, glass, soil, sludge, scrap metal, insulation materials, etc.). Total waste humidity may reach 40% with individual packages containing up to 90% of water. Plasma

method can process in a shaft furnace such inconvenient for other high-temperature technologies waste as ion exchange resins, activated carbon and inorganic sorbents discharged from the drain water purification devices and liquid radioactive waste, and characterized by high moisture content. Previously packed in barrels or supercompacted waste were successfully processed on the "Pluton."

The organic part of the radioactive waste is subjected to pyrolysis in the shaft furnace under lack of oxygen while melting of the slag is carried out in an oxidizing atmosphere that promotes the complete destruction of the organic constituents of the slag, and gives a more uniform product.

NPP Radioactive Waste

According to the Russian "Law on the Use of Atomic Energy" (November 21, 1995 N170-FZ) radioactive waste, is nuclear materials and radioactive substances, without planned further use. Often they are the products of nuclear processes, such as nuclear fission. Most of the radioactive waste are so-called low-activity wastes with low radioactivity per mass or volume. For example, used protective wear, slightly polluted, but still representing a danger of radioactive contamination of the body through the pores of the skin, respiratory tract, water or food.

During NPP operation in addition to the fuel cycle products produced in fission reactions, byproducts that are not related to the fuel cycle, but having a residual activity also formed. These include construction debris, insulation, paper and other items that were in contact with radioactive materials and components. The composition of such waste is presented as an example of solid radioactive waste from Novovoronezh NPP[2] (Table 1, 2).

Table 2. Radionuclide composition of solid low- and intermediate active waste from Novoyoronez NPP

Radionuclides	Total activity fraction, %	Specific activity of solid waste, Bq/kg	Specific activity of used ion exchange resins, Bq/kg
Cs-137	54.0	7.94×10^5	7.02×10^6
Cs-134	4.5	6.62×10^4	5.85×10^5
Co-60	22.0	3.23×10^{5}	2.86×10^{6}
Co-58	1.5	2.2×10^{4}	1.95×10^{5}
Sr-90	13.0	1.9×10^{5}	1.7×10^{6}
Mn-54	5.0	7.4×10^4	6.5×10^{5}
Total	100.0	1.47×10^{6}	1.3×10^{7}

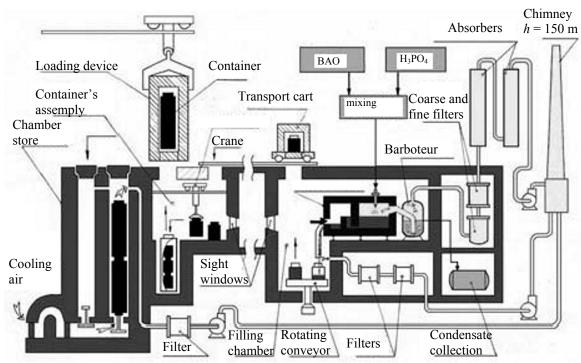


Fig. 1. Vitrification of liquid waste on EP-500 unit at PO "Mayak" (Russia).

Traditional Methods of Radioactive Waste Processing

According to the adopted in Russia concept complexes for processing of solid radioactive waste from NPP and nuclear technological cycle plants production line include pretreatment of solid waste for recycling (waste retrieval from storage, their fragmentation and sorting), waste thermal processing, in particular, solid and liquid combustible waste burning plants with the ash cementing assembly, thermal insulation melting units.

Conditioning of solid radioactive waste, i.e. transforming them into a form suitable for safe storage and transportation is carried out by compression and supercompacting methods. Packages with compacted and cemented waste are placed on the long-term storage in non-reusable protective containers, that also used for storage some of the waste without processing. These "cold" radioactive waste conditioning methods are characterized by relatively low coefficients of reducing waste volume, therefore requires a large area, and bulk storage to accommodate conditioned waste forms [3].

Combustible solid radioactive waste is processed at the burning facility, followed by ash conditioning by cementing. Mixed solid waste that can not be incinerated, sent sequentially to the pre pressing in 100 L barrels and subsequent supercompacting (placed in 100-L barrels) in supercompactor. Conditioned waste are placed in barrels (200 L) in non-reusable protective container

More rational way of conditioning of radioactive waste is vitrification. There are two major waste vitrification process: with calcification before melting and without calcination. Conventionally, they are called two-stage and single-stage processes [4]. Two-stage process is used in France and the UK, single stage, in the U.S., Russia, Germany, Belgium, Japan, India, South Korea.

Vitrification starts with the evaporation of the water, after which the concentrate is calcined (in a two step process), or immediately sent to the mixing with the glass forming additives (in a one step process). As additives in the two-step process is used in the form of glass beads (so-called cullet) in a one-step process - sand, clay, natural borated datolite, and other materials which improve the pulping process and the properties of the final product. A mixture of radioactive waste from glass-forming additive is supplied into the furnace for radioactive glass melting. This can be a big bathroom oven, like in the window glass manufacture, or induction furnace. Heating the bath to the glass melting is carried out either by passing current through

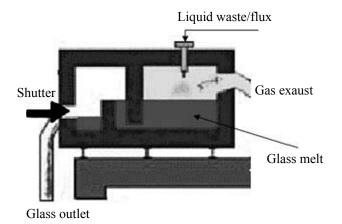


Fig. 2. Electrical melter EP-500 for vitrification of liquid active waste (PO "Mayak").

molten glass (such a method is used in Russia), or by inducing high frequency currents by external inductor.

Figure 1 gives a flow diagram of the process of vitrification of liquid radioactive waste in the Russian "Mayak" plant, melter EP-50 scheme is shown in Fig. 2.

Table 3. Radioactive waste processed on "Pluton" [8]

Component	Content, wt %
Paper	11–90
Wood (saw dust)	1–5
Wood (fire wood)	2–20
Textile (clothings)	4–7
Plastics (PE, PVC)	4–8
Glass (domestic and laboratory)	2–8
Rubber (tubings, tires)	2–5
Electronic components	1–5
Construction waste	4–15
Thermal insulation (glass- and rockwool)	1–5
Metal	3–10
Ion exchange resins	0.3–2
Plants materials	2–5
Total ash content	7–40
Total moisture content	5–24

On the example of EP -500 can be seen that for vitrification of radioactive waste according to the scheme considered requires expenses for primary processing of waste, filtering flue gases and the introduction of components for vitrification. Costs are at least partially reduced in the case of plasma processing of radioactive waste.

Plasma Processing of Radioactive Waste in Russia

The first full-scale plasma system for radioactive waste treatment, ZWILAG in Switzerland, was commissioned in 2004. The maximum capacity of the plant is 200 kg/h of combustible waste and 300 kg/h of non-combustible meltable waste [5].

The second full-scale industrial system for plasma processing of solid radioactive waste was the "Pluton," commissioned by NPO "Radon" in Russia in 2007 [6, 7].

Pilot plant "Pluto" was designed for processing of radioactive waste of mixed type (Table 3). Processing technology based on high-temperature plasma pyrolysis of waste in flow blast furnace to obtain slag compound, extremely resistant to aggressive environmental influences. Performance of the unit is up to 200–250 kg/h.

Flow diagram of "Pluton" is shown in Fig. 3.

Shaft furnace of the unit is made of refractory and insulating materials with exterior cladding with steel sheets. Shaft height is 6.4m (from the floor), the internal cross-section -0.8×0.8 m, capacity 3.5 m³. In the vault of the melting chamber there are two plasma torches of 100-150 kW each, which provide a melt temperature of $1500-1700^{\circ}$ C.

Waste to be processed through the vane load device introduced into the upper part of the shaft. By gravity feed material moves down the shaft, wherein it is heated by the heat of the exhaust gas moving upstream material in the shaft furnace. In the upper and middle layers of the shaft waste is drying due to the heat of exhaust gases and undergoes pyrolysis in the absence of free oxygen. Organic residues and refractory inorganic constituents enter the lower layers of the shaft, the coke combustion and slag melting zone.

Molten slag accumulates in the melting bath, where it is homogenized, overheating and over discharging vane enters the slag filling chamber. From there the slag is discharged in a continuous or batch mode in

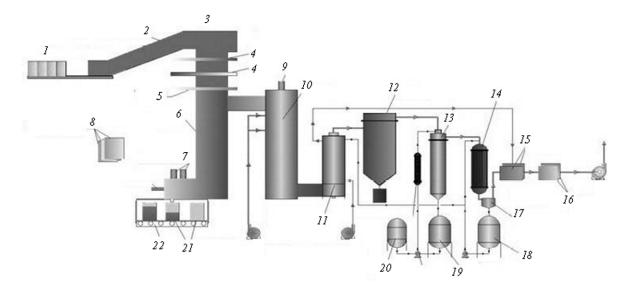


Fig. 3. "Pluton" technological scheme [5]: (1) feed chamber; (2) hermetic conveyor; (3) loading chamber; (4) locks; (5) heat shield; (6) shaft furnace; (7, 9) plasma torches; (8) plasma torches power supply; (10) afterburning chamber; (11) heat exchangers; (12) bag filter; (13) scrubber; (14) condenser; (15) gas mixer; (16) fine filter; (17) gas separator; (18) condensate collector; (19) circulating tank; (20) alkaline dispenser; (21) metal containers; (22) melted slag filling chamber.

metal containers. The temperature of the slag melt in the bath furnace reaches 1600–1800°C, while the temperature of flue gases at the outlet of the shaft furnace is not higher than 250–300°C. After cooling of the melt in the receiving container solidified slag is sent to landfill for long-term storage of conditioned radioactive waste.

It is possible to supply the shaft with blast air to regulate the performance of the furnace or pyrolysis. Pyrolysis gases from the kiln shafts come in a afterburning chamber, where combustible gas and aerosol components of pyrolysis burn at 1100–1300°C.

Further, the exhaust gases are cooled in evaporative heat exchanger to 300°C, cleared of aerosols in a bag filter and cooled in a heat exchanger. Noxious gaseous components (HCl, NO₂, SO₂) is neutralized in the absorber, by circulating alkaline solution. Before release into the atmosphere of the exhaust gases pass additional sanitary cleaning in absolute filter.

For furnace heating arc plasma torches are installed in the bottom of the furnace above the bath, using air as the plasma gas. For furnace and afterburner heating DC plasma torches with electric power of 100–150 kW developed by NPO "Radon" are used.

Unit is equipped with sensors, monitoring temperature, pressure, electrical parameters and flow environments. Operation of the unit is made from the control board that hosts the instrumentation, plasma torches current controls, complex for registration and control of technological parameters on the base of the computer and controller, start and emergency shutdown of the plasma torches buttons and status indicators.

Plasma processing of radioactive waste produces slag compound to be buried. Table 4 shows the composition of the slag, chemical and radionuclide, produced by "Pluton."

Glass-like slag compound matrixing active elements, practically is not subject to leaching of these elements. The volume of the slag compound is 20–50 times smaller than the volume of the raw waste. Buried slag compound is significantly less risk in comparison with the disposal of radioactive waste in concrete blocks, since the probability of getting radioactive substances into the human body through the food chain and water is significantly lower. At the same time considerably less the volume of waste to be disposed of.

Radioactive slags are extremely resistant to chemical attack, the rate of leaching of radionuclides (¹³⁷Cs) in water in average is 10 times lower than for borosilicate glass and is located at 10⁻⁶ g cm⁻² day⁻¹ level. The density of the obtained slags is 2.7–2.9 g/cm³, the mechanical strength of 600 to 800 MPa. In its properties, radioactive slags from the "Pluton" are similar to the volcanic glasses.

Table 4. "Pluton" slags composition

	•	-					
Al_2O_3	SiO ₂	Na ₂ O	K ₂ O	CaO	MgO	Fe ₂ O	ZnO
Chemical composition (in wt %)							
18.8-27.9	35.0–56.0	2.6-11.1	0.6-2.1	2.1-8.7	1.2-2.9	1.5-8.5	0.1–13
Cl	P_2O_5	S	B_2O_3	PbO	NiO	Cr ₂ O ₃	CuO
0.1-0.3	0.3-0.7	0.06-0.1	0.7-3.7	0.3-4.1	0.1-3.7	0.2-0.6	0.1–3.1
Radionuclide composition (in Bq/kg) ^a							
$\sum \! eta_{rel}$	¹³⁷ Cs	$\sum \! lpha_{rel}$	²³⁹ Pu	²⁴¹ Aı	m		¹³⁷ Cs
1.1×10^{4}	-1.3×10^6	$10^6 5.2 \times 10^3 - 6.8 \times 10^6$		$5.0 \times 10^3 - 2.7 \times 10^4$		$2.7 \times 10^3 - 1.7 \times 10^5$	
134	Cs	⁹⁰ Sr+ ⁹⁰ Y ⁶⁰ Co)		²²⁶ Ra	
7.5×10^{2}	$7.5 \times 10^2 - 1.3 \times 10^4$ $2.1 \times 10^3 - 2.8 \times 10^5$ $5.0 \times 10^2 - 3.0 \times 10^3$		38–590				
239	239 Pu 238 Pu 238 U		²³⁵ U	^{234}U			
1.8×10^4	-5.9×10^5	9.2×10^{2}	-2.5×10^5	$1.2 \times 10^3 - 5$	5.7×10^3	50–210	$2.8 \times 10^3 - 1.4 \times 10^4$

^a Maximal slag specific activity 7×10^5 Bq/kg a-radiating and 1.1×10^7 Bq/kg β -radiating isotopes.

Table 5. "Pluton" flue gas composition. Flue gases content 2000 kg/h from total 55000 kg/h of ventilating system. Gas temperature 25°C

Component	Concentration	Emission, kg/h
H ₂ O	15 g/m ³	40
H_2	0.15 mg/m^3	0.01
СО	3 mg/m^3	0.2
SO_X	0.5 mg/m^3	0.03
NO_X	0.2 mg/m^3	0.01
HCl	0.015 mg/m^3	0.001
C_XH_V	0.15 mg/m^3	0.003
Resins	0.015 mg/m^3	0.001
Dust	0.015 mg/m^3	0.001
Polycyclic aromatic hydrocarbons (PAHs)	$0.004 \ \mu g/m^3$	0.3 mg/h
Polychlorinated dioxins and dibenzofurans (dioxins)	0.00 4 ng/m ³	0.1 μg/h
α-Emitting nuclides	0.004 Bq/m^3	300 Bq/h
β-Emitting nuclides	0.07 Bq/m^3	5000 Bq/h

The resulting flue gases contain, along with radioactive aerosols, inorganic pollutants and organic toxicants (Table 5). As "Pluton" operating practice shows, it has less impact on the environment compared with the incineration chamber furnace.

As a result of R&D carried out by NPO "Radon" in cooperation with NPO "Typhoon," it was found that concentration of polychlorinated dibenzo-p-dioxins and dibenzofurans (hereinafter dioxins) in terms of the toxic equivalent (TEQ) in the pyrolysis gas at the

outlet of the plasma shaft furnace on average are five times less than in the flue gas of chamber furnace incinerator. Total content of dioxins in the flue gas at the outlet of "Pluton" gas cleaning system does not exceed 0.014–0.02 ng/m³ TEQ, which is about five times lower than the European standard for waste incinerators. Concentration of heavy metals in Industrial gas emissions into the atmosphere is also below standards for waste incinerators in Western Europe.

Prospects for Implementation of Plasma Processing of Solid Radioactive Waste in Russia

This section provides a brief overview of the development of plasma technology for radioactive waste treatment using recycling complex of Novovoronezh NPP in Russia [9, 10] as example.

State Corporation "Rosatom" in 2008 decided to establish a technology demonstration center for radio-active waste management on the basis of Novovoronezh NPP where simultaneously operated three power units, decommissioned two blocks and is building two new power units of the second stage of the nuclear power plant. Various technological facilities for processing and conditioning of radioactive waste are planned to place there. Plasma processing complex for solid waste of low and intermediate active level, which is currently under construction, will be one of the most important objects of an industry demonstration center.

Over 30000 m³ of solid waste of low and intermediate level are accumulated at Novovoronezh NPP, in addition, about 380 m³ of waste enters the store annualy.

The performance of the plasma processing complex of solid radioactive waste is 1750–2000 m³ per year. The complex provides a reduction in waste volumes by 30–40 times, which is an average of approximately 50 m³ per year of waste that go to storage in conditioned form. Thus, the operation of the complex will annually release 1700–1900 m³ of radioactive waste storage.

In most, technical options for plasma waste processing facility of Novovoronezh NPP (Table 6) are the same as for "Pluton," but there are some differences (weight not more than 10 kg, size limits for any section of a single subject in the composition of radioactive waste 300 × 300 mm).

Plasma reactor is a complex lining apparatus having a cooled outside elements. Waste loaded from above

Table 6. Technical parameters of radioactive waste processing facility at Novovoronez NPP

	I
Process	Value
Radioactive waste performance, kg/h	200–250
Slag performance, kg/h	50-80
Pyrogas performance, kg/h	300–400
Flue gas performance, m ³ /h	2000–3000
Temperature in melter, °C	1500-1800
Pyrogas temperature, °C	up to 300
Plasma torches electrical power, kW	2 × 100–150
Afterburning chamber plasma torches electrical power, kW	50–60
Total electrical power, kW	850
Waste feed method	In kraft sacks

through the lock system to prevent air leaks into the reactor and the output of refined products outside.

The overhead area of the lower portion of the reactor contains plasma torches, a device for air blust, steam (if used) and other necessary components as well as a device to output the molten inorganic material. Air is used as plasma-forming gas.

In the reactor the waste pass several transformations zones, namely zones of drying, pyrolysis, gasification, burning, melting and vitrification. The complex also includes pyrogas afterburner, the gas cleaning compartment (two redundant lines and explosive compensation valve system), including evaporative heat exchanger, scrubber, a flue gas cooling system, the flue gas filtering unit, and a control system for sampling gas emission.

When operating in the normal mode, the complex outputs slag compound waste that is loaded in concrete blocks, followed by transporting the containers for long-term storage in the storage of solid radioactive wastes and radioactive waste gases emitted during post-combustion of the pyrolysis gas as well as general ventilation work premises. In addition, it produces salt solutions (law level watering), forwarded to the existing special drainage of first and second reactors

for further processing, and dry residue that sent for processing in a plasma reactor.

The complex will be located at the site of NPP within the sanitary protection zone established by applicable laws and rules.

Radiation situation on the territory, the type and content of radioactive substances in the environment (air, vegetation, soil and dose level on the ground), will be determined by emissions of gases and aerosols in surface air from the ventilation systems of nuclear power plants and the complex' chimney.

Project includes three flue gas cleaning systems. Ventilation air from partly attended and unattended premises before release into the vent pipe must be cleaned on the filters. According to the draft, gas cleaning system provides a degree of purification of not less than 10^3 ; specific activity released into the air vent pipe is not more than 3.7 Bq/m³, well below the allowable level. Total gas and aerosol emissions into the atmosphere during normal use will not exceed 4.9×10^4 Bq/day or 1.5×10^7 Bq/year, which is about 1% of the annual allowable emissions. Thus, the radiation doses to the population due to the release of radionuclides from the proposed facility will not exceed the established limits.

Sources of radioactivity in the operation of the complex in normal mode are equipment containing radioactive substances (liquid radioactive waste containers, mixers, filters, pumps, containers with slag compound) and gaseous radioactive waste after pyrolysis of solid waste in the shaft furnace, as well as emissions from the premises contaminated air supply and exhaust ventilation.

Safety system of the facility is based on the concept of multi level protection, based on the use of physical barriers to the spread of ionizing radiation and radioactive substances into the environment, to implement technical and organizational measures to protect and preserve the barriers of their effectiveness, and to protect workers, the public and environment.

System of physical barriers for radioactive waste and by-products include leaks proof equipment, its biological protection, leakage collecting tray, matrix cemented waste transport container that serves as the localization and biological protection. Estimated population exposure dose during normal operation of the complex on the border of the existing sanitary protection zone (5 km) is less than 0.1 mSv/year. Project personnel exposure dose limit set at design accident is assumed to be 20 mSv/year. Design dose limit for public exposure during design accident at the border of the sanitary protection zone is 1 mSv/year but for beyond design basis accidents at the border zone of planning protective measures, 5 mSv/year. To control for the specified parameters radiation monitoring system is provided.

Release of radioactive substances during normal operation of the complex into the environment is only possible through the pipe with the exhaust air. Ventilation air before release into the pipe is subjected to multistage cleaning on filter. The specific activity of air discharged into the pipe does not exceed the permissible volume activity for the population according to NRB-99. By repeated dilution air in the surface layers of the concentration of radioactive substances will be three or more orders of magnitude lower than the original, that would not change the radiation situation in the environment

Thus, under normal use and in the event of an accident, personnel individual doses will not exceed the specified limits, and getting into the environment radioactive substances create radiation dose of the population by several orders below acceptable.

CONCLUSIONS

The introduction of plasma technology for the processing of solid radioactive waste of nuclear power plants increases the economic efficiency of radioactive waste processing due to economies on waste storage space and reducing of waste storage equipment and operations of waste conditioning.

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